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Title:	Irradiation induced changes in small angle grain boundaries in mosaic Cu thin films
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Irradiation induced changes in small angle grain boundaries in mosaic Cu thin films

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CMIME Center for Materials at Irradiation and Mechanical Extremes

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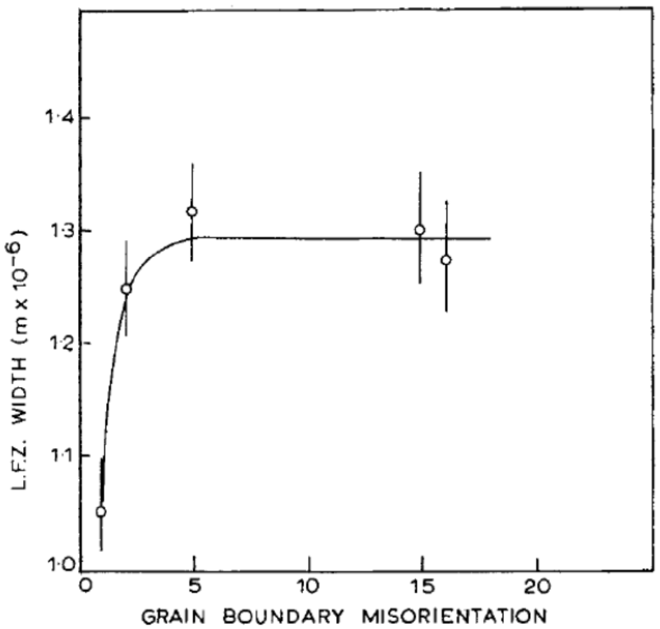
Outline

- Motivation
- Material and characterization method
- Properties of as-deposited films
- Change in properties of films after ion irradiation
- Potential mechanism

Motivation

- **Several groups reported the efficiency of grain boundaries as sinks for point defects is depending on grain boundary mis-orientation.**



The effect of boundary mis-orientation on loop free zone (L.F.Z.) width in oil-quenched Al-1.5%Zn

J. Burke, et al, Phil. Mag. 31 (1975) 1063.
B.K. Basu, et al, Acta Metall. 13 (1965) 117.
J.D. Embury, et al, Acta Metall. 13 (1965) 403.

Motivation

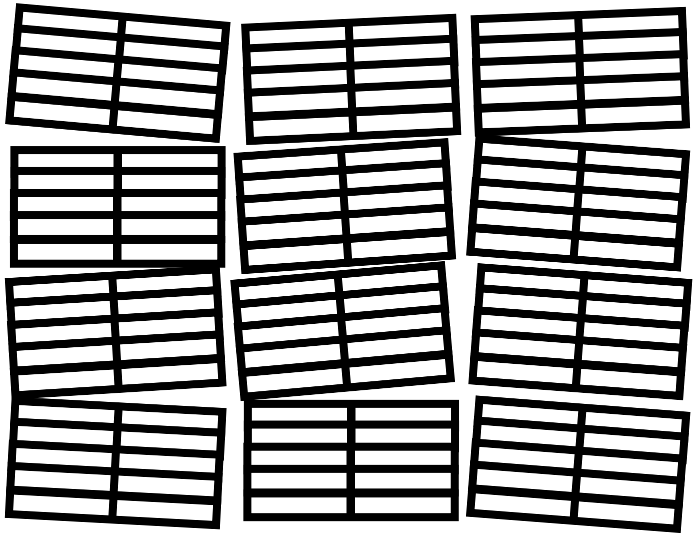
- **Several groups reported the efficiency of grain boundaries sinks for point defects is depending on grain boundary mis-orientation.**
- **The change of small angle grain boundaries after absorbing the point defects is not well reported and understood.**
- **Questions:**

  (1) How do small angle grain boundaries interact with irradiation induced defects?

  (2) Are small angle grain boundaries still effective sinks for irradiation produced defects?
- **Study: Interactions between interstitials and small angle grain boundary with mis-orientation below 1 degree.**

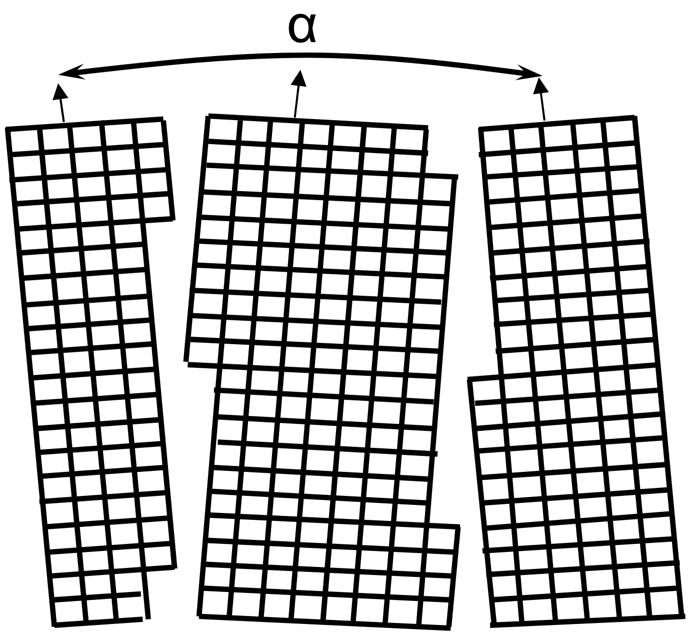
Mosaic Crystal

- The notion of mosaic structure was first introduced by Darwin to describe the microstructure in single crystals.
- A single crystal is made up of small perfect-crystal blocks, each slightly mis-orientated one from another.
- Block size: the order of 100 nm and the maximum angle of disorientation between them from very small to one degree.

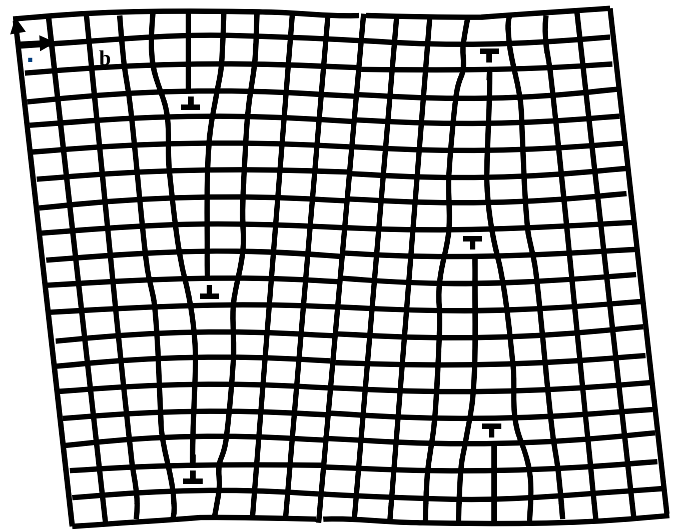


C.G. Darwin, Phil. Mag. 27 (1914) 675.

Model of mosaic crystal



Model of mosaic crystal



Sub-grain with small angle GB

- Mosaic crystal is composed of sub-grain with small angle mis-orientation between them
- Small angle grain boundaries become bridges to connect each block with its neighbors. The small angle grain boundary is comprised of an array of dislocations.

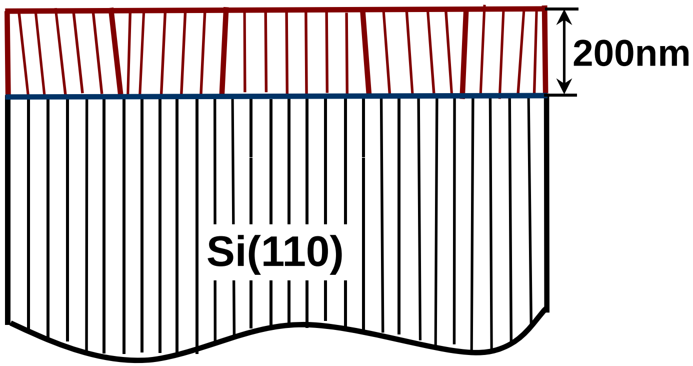
Materials and methods

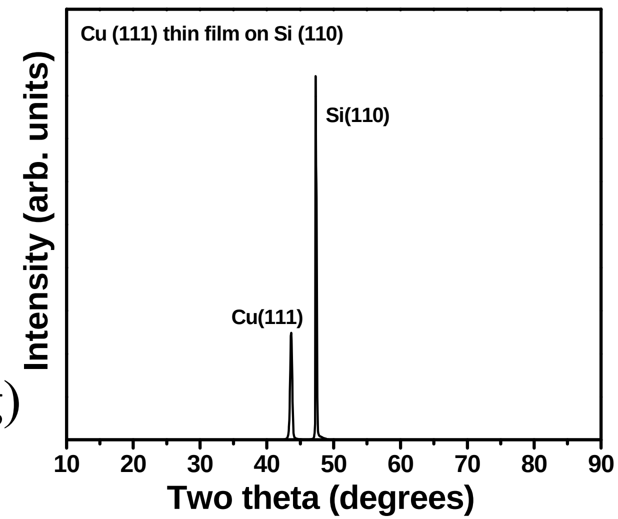
- **Synthesis of Cu films with mosaic structures**

  1. Magnetron sputtering
  2. HF etched Si(110) substrate
  3. Room temperature
  4. Ar pressure: 4 mTorr
  5. Total thickness: 200 nm

- **Characterization of Cu films**

  1. Rocking curve of X-ray diffraction (XRD)
  2. RBS analysis (Random and Channeling)





Methods to examine mosaic spread of thin film

- **Method 1:**

  Rocking curve of XRD was used to measure the mosaic spread of film

- **Rocking curve:**

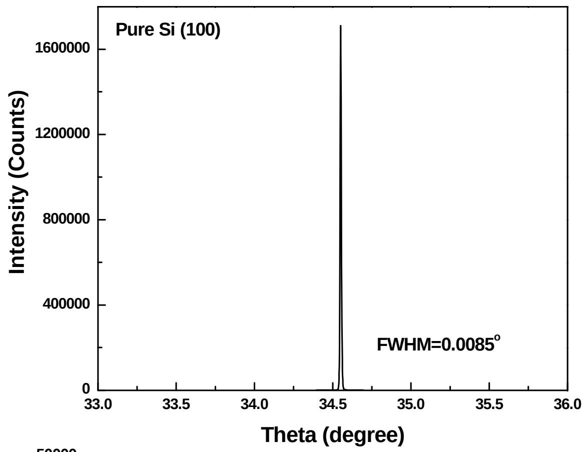
  The angle (θ) and the detector position (2θ) is fixed at the Bragg angle of the corresponding reflection. Rocking curve is acquired by rotating sample through the Bragg angle.

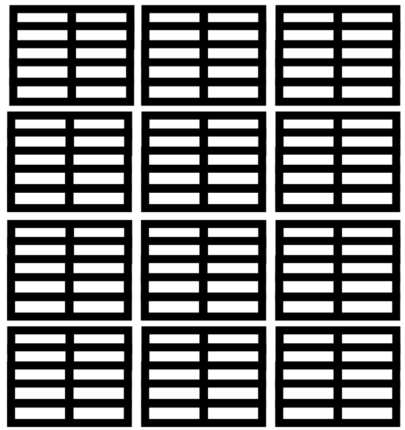
  Mis-orientation angle α and incidence angle θ, then diffraction at all angles between θ and $\theta + \alpha$

- **Information from Rocking Curve:**

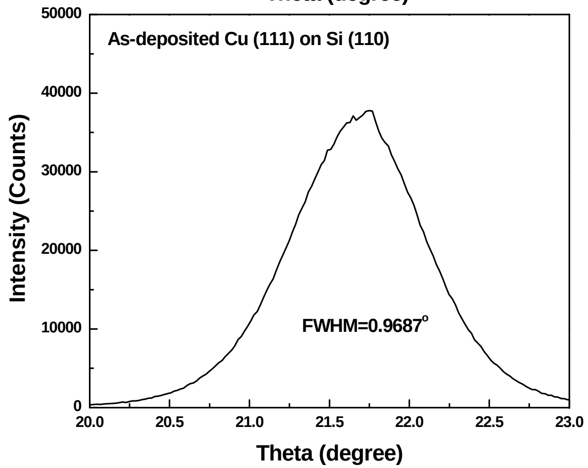
  Full Width at Half Maximum (FWHM) of rocking curve determine the mean spread of mosaic crystal

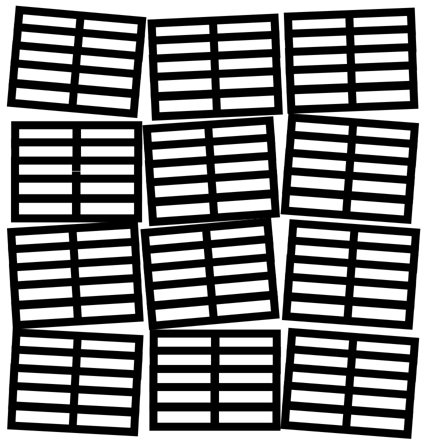
Comparison of rocking curves between Si (100) and as-deposited Cu(111)/Si(110)





Perfect crystal



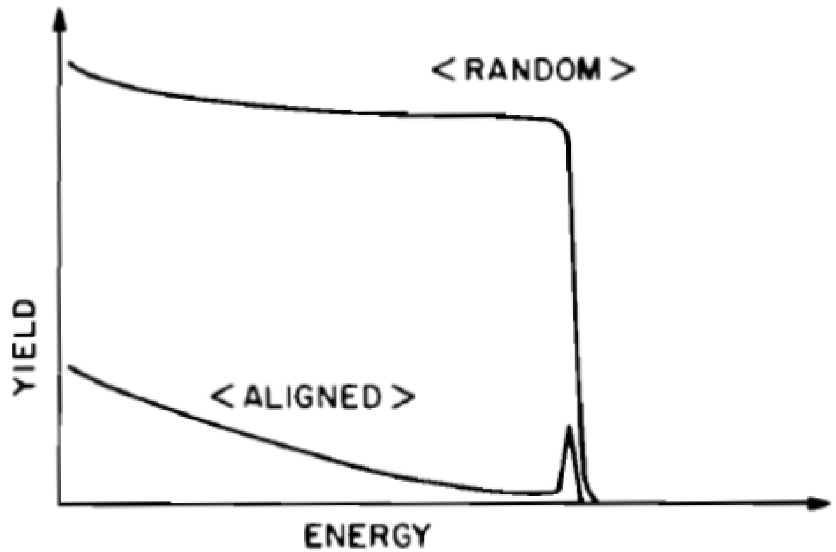


Mosaic crystal

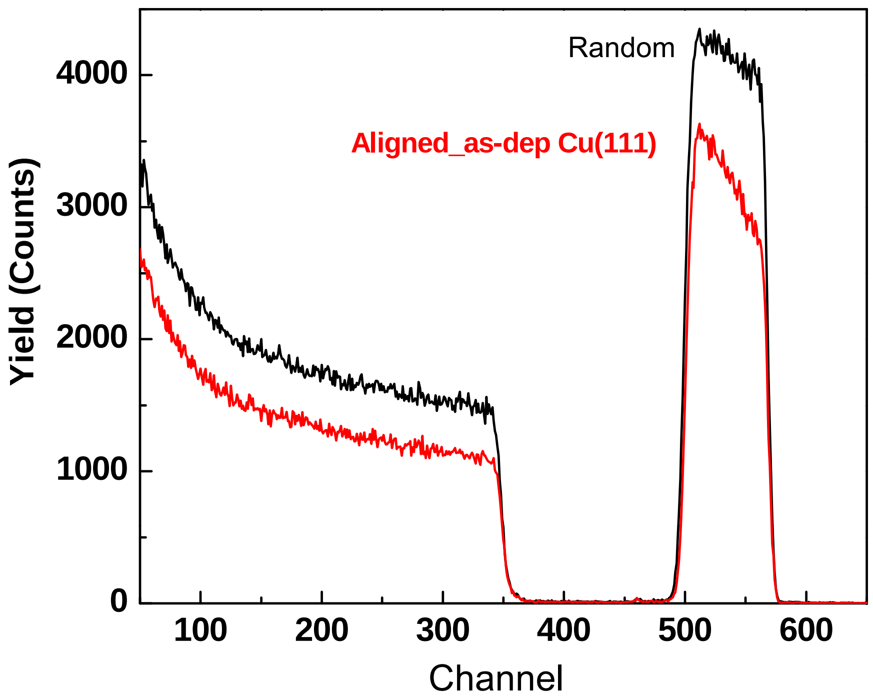
- Two rocking curves are over the same range of θ
- Comparison shows peak breadth of Si (100) is much narrower than that of Cu (111), indicating large mosaic spread in Cu.

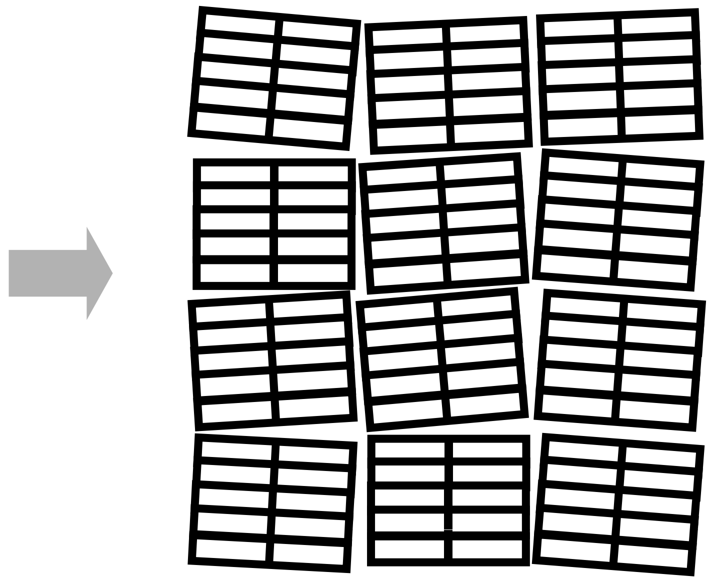
Methods to examine mosaic spread of thin film

- **Method 2: Rutherford backscattering spectrometry (RBS):** determine the structure and composition of materials by measuring the backscattering of a high energy ion beam impinging on a sample.
- **RBS channeling:** Strikingly large reduction in the yield of backscattered particles as the orientation of the single crystal target is aligned with the incident beams.
- **Minimum Yield:** The ratio of the heights of two spectra taken in the near-surface region for aligned and random orientation



RBS/C spectra of as-deposited Cu film





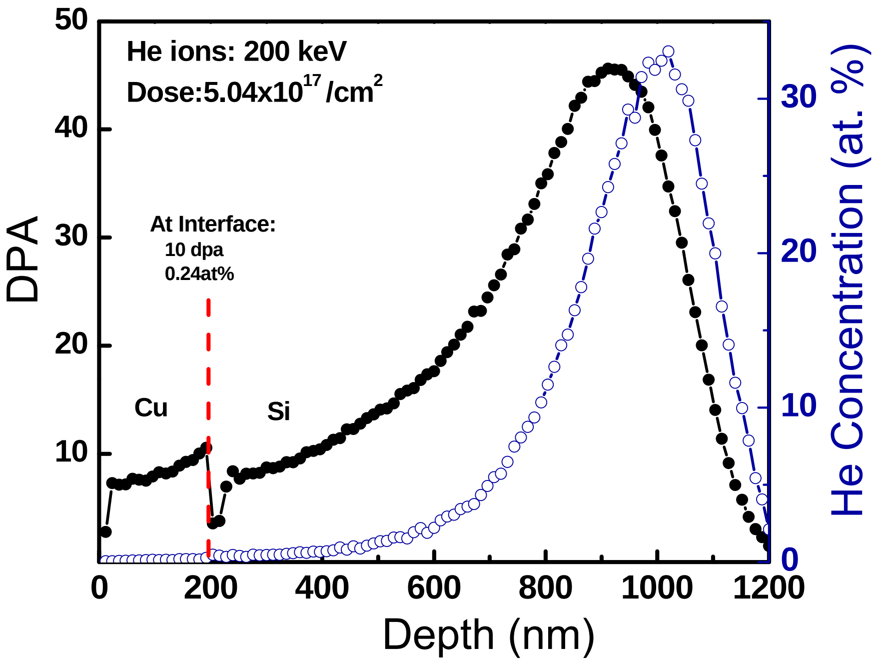
Mosaic crystal

- The random and channeling spectra of RBS measurement for as-deposited film shows the minimum yield of 70%, indicating mosaic crystal with small misorientations in the film.

J.F. Ziegler, M.D. Ziegler, J.P. Biersack, http://www.srim.org

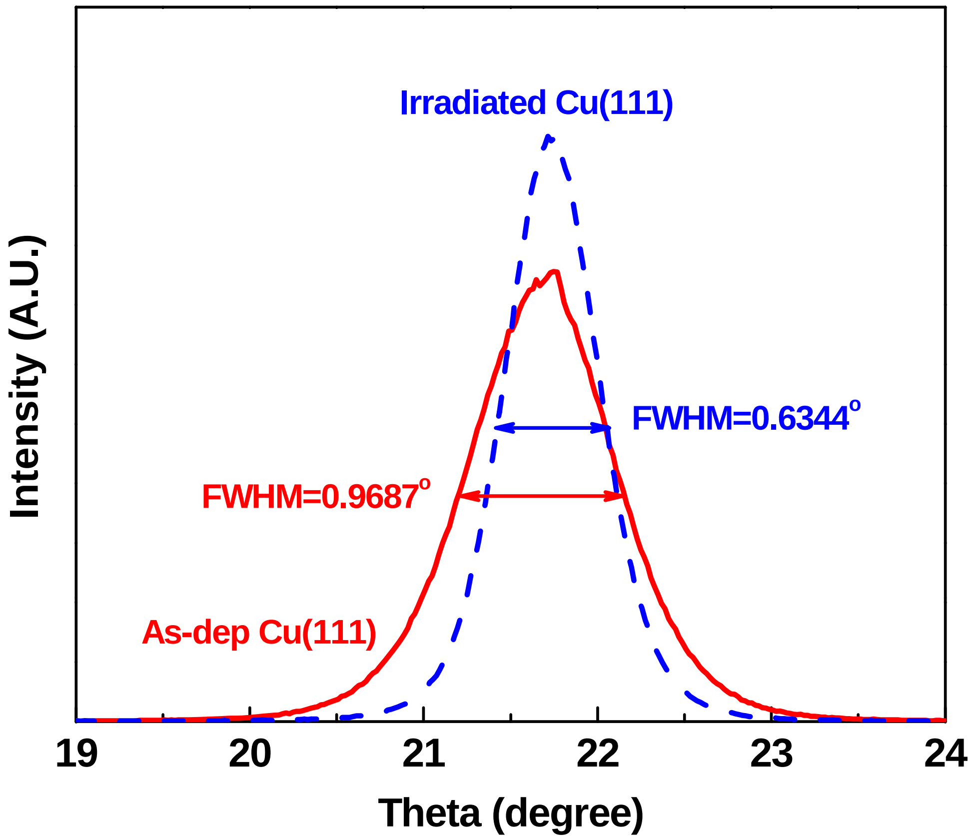
Helium ion irradiation experiment

- **Purpose:** Radiation response of mosaic structure in Cu film
- **Concern:** He bubbles in the films (expect ultra low ratio of He bubbles to damage)
- **TRIM simulation**



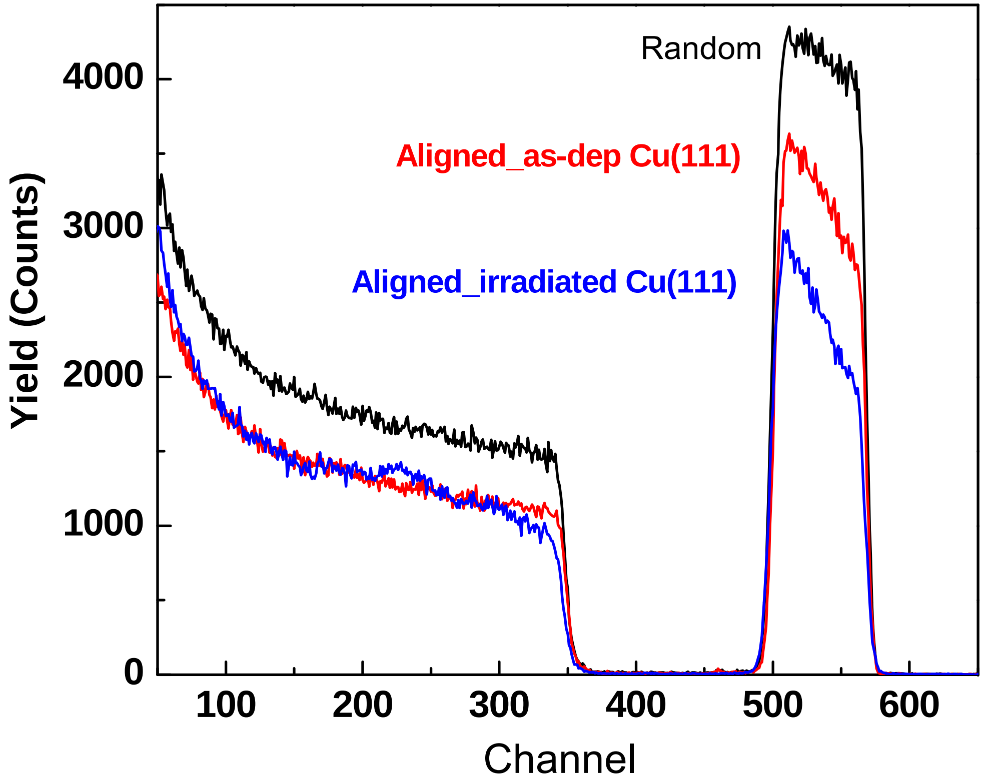
- **Helium radiation:** 200 keV; 5×10^{17} ions/cm^2; room temperature.

Rocking curves of Cu (111) in as-deposited and ion irradiated Cu (111) film on Si (110)



- Comparison shows the FWHM of rocking curve decreases 35 % after ion irradiation with fluence $5\times10^{17}/cm^2$, indicating 35% decrease of mosaic spread.

RBS/C spectra of as-deposited and ion irradiated Cu films with fluence 5×10^{17}/cm^2



- Comparison shows minimum yield decreases 30% after ion irradiation with fluence 5×10^{17}/cm^2, indicating 30% decrease of mosaic spread.

1. Mobility of interstitial and vacancy

- **Diffusion coefficient of point defect:**

$$D = \alpha a^2 v \exp(-E_m / kT) \exp(S_m / k)$$

where alpha is a constant, a is the lattice constant, v is frequency, S_m is entropy, and E_m is migration energy, for interstitial is 0.12 eV and for vacancy is 0.8 eV in Cu.

- **The ratio of diffusion coefficient of interstitials to vacancies at RT:** 2.8×10^{11}, so at low temperature (RT), the interstitial is mobile, and the vacancy is immobile.

G.S. Was, Fundamentals of radiation materials science: Metals and Alloys, Springer Berlin Heidelberge New York, 2007.

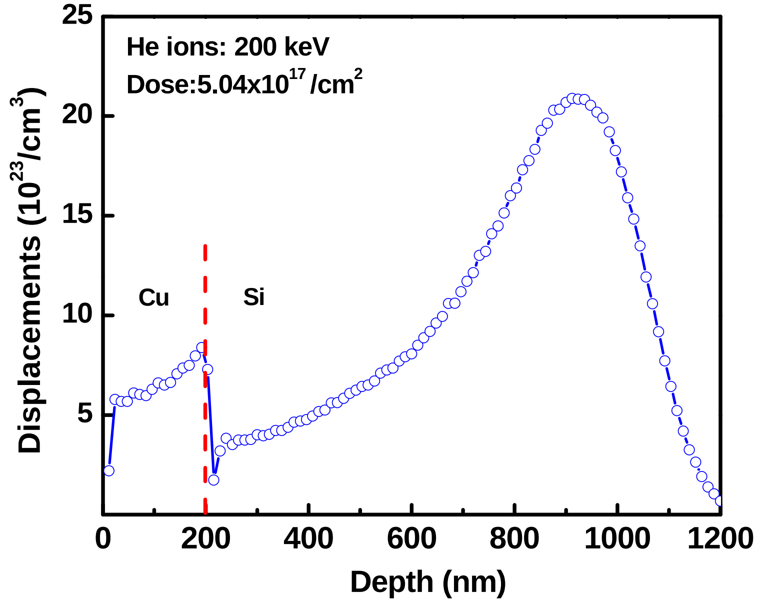
2. Point defect concentration of as-deposited Cu film

- **Equilibrium interstitial concentration in bulk Cu:** $N_v^0 = N \exp(\frac{-u_v}{kT})$

where u_v is formation energy, T is temperature, so:

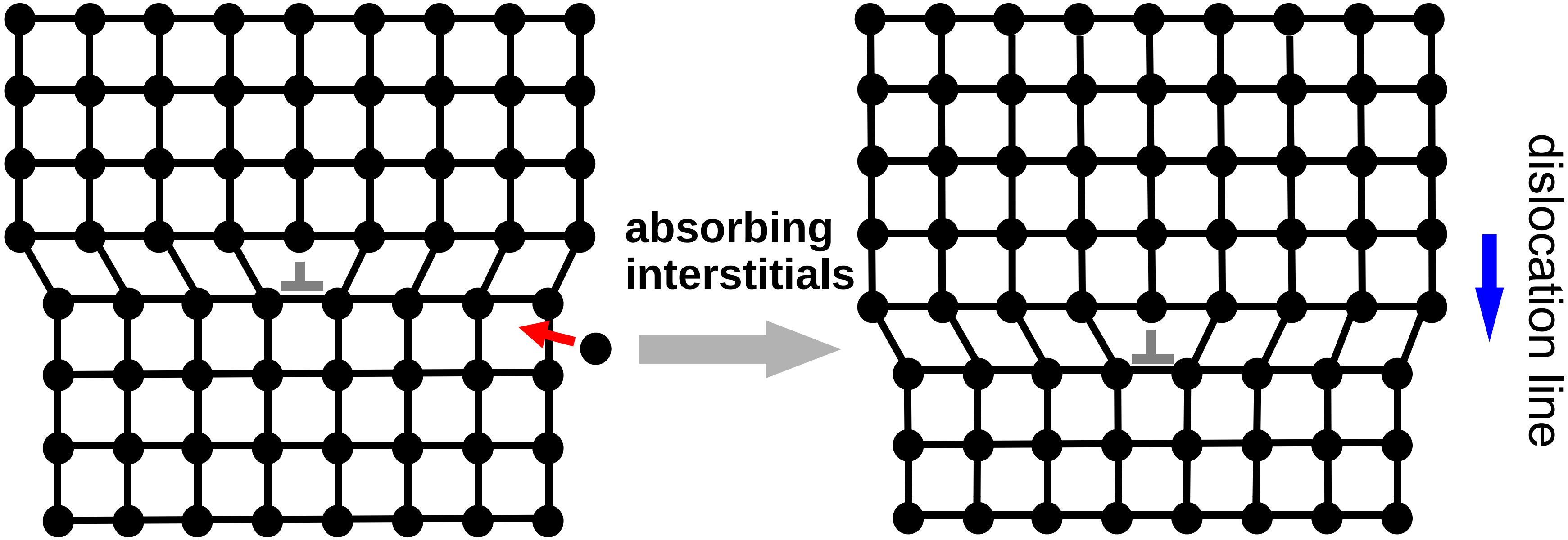
Equilibrium concentration of interstitial = **7.8×10⁻¹⁵**/cm³

- **Defect concentration after irradiation:** Displacement concentration after irradiation is $\mathbf{7.5\times10^{23}/cm^3}$ at $5\times10^{17}/cm^2$. The fraction of defects that are freely migrating after quench stage is **0.02**, so the concentration is $\mathbf{15\times10^{21}/cm^3}$, much higher than interstitial concentration in as-deposited film.



Callister W.D. Jr., Materials Science and Engineering: An Introduction, 5th ed. John Wiley & Sons, Inc. 2000

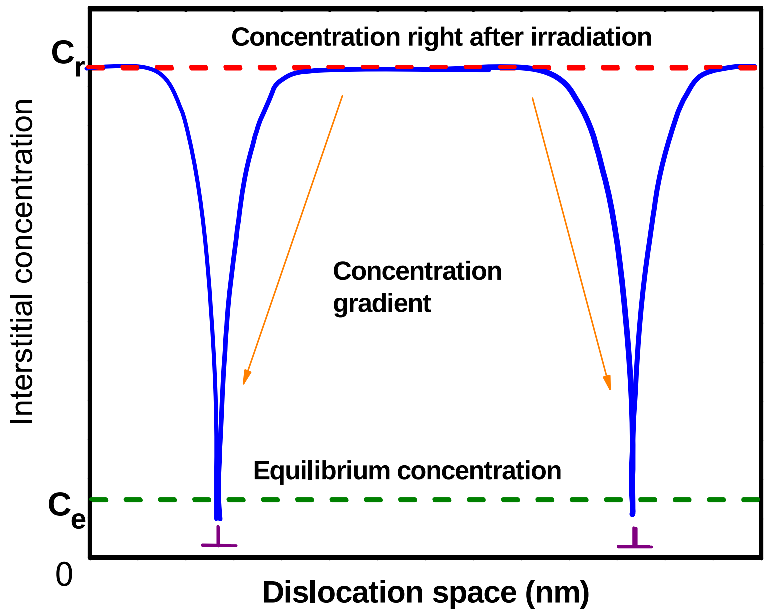
3. Dislocation negative (down) climb by absorbing interstitials



- Dislocation negative climb by absorbing interstitials under diffusion control.
- Dislocation line moves down by dislocation negative climb

4. The creation of concentration gradient in the region close to dislocations

- **Before irradiation:** Point defects with low concentration equilibrate at dislocations because dislocations can act as sources sinks for point defects in the as-deposited sample. The dislocations motions cease.
- **After irradiation:** at the start, the radiation induced interstitials uniformly distributed but with much higher concentrations, which is constant and uniform in space
- **Creation of concentration gradient:** Much higher interstitials concentration breaks the balance close to dislocation. So dislocation climb by attracting the interstitials, which reduces the local concentrations of interstitials again. As a result, the concentration gradient is produced



Conclusions

- **As-deposited Cu film on Si substrate has a mosaic structure.**
- **Rocking curve measurements show that He ion irradiation reduced FWHM.**
- **RBS data show a reduction in minimum yield after irradiation**
- **Both evidences indicated mosaic structure is improved by ion irradiation.**
- **Reduction in mosaic structure results from interstitial absorption at small angle grain boundaries by dislocation climb in mosaic structure.**
- **Small angle grain boundary angle and mis-orientation angle decrease after ion irradiation**